Geometrical model for martensitic phase transitions: study of the effective dipole-dipole correlation between neighbor domains.

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Abstract: A model for the growth of martensitic domains during a structural phase transition is studied by numerical simulation. As a consequence of the absence of retransformation mechanism, the domains in the final microstructure show an effective dipole-dipole correlation, increasing the probability of finding neighboring parallel domains. We have focused our attention on a two-dimensional case in which two perpendicular symmetrically-related domains grow with, a priori, equal probability. The attractive correlation is shown to decay logarithmically with the perpendicular distance between parallel domains.

I. INTRODUCTION

Martensitic transformations are displacive (without atomic diffusion) solid-solid first-order phase transitions in which the symmetry of the crystalline lattice changes due to a shear mechanism. This involves a displacement of the atoms typically smaller than the interatomic distances [1, 2]. The high temperature phase is known as austenite phase whereas the low temperature, less symmetric phase is called martensitic phase. This transformation is crystallographically reversible but with a certain hysteresis in the transition temperature [3]. When cooling or stressing, the phase transition occurs by nucleation of the martensitic phase in the austenite phase, creating a certain microstructure. This microstructure is determined by the natural tendency of systems to keep their minimum energy state. In order to minimize the elastic strain field, the martensite can grow in multiple symmetry-related and energetically equivalents variants. In the simplest example, starting from a square lattice in 2D the system can transform to two different rectangular martensitic domains rotated 90°. Due to the elastic energy minimization process, the transformation of a certain region can induce the transformation of their surrounding atoms in an auto-accommodation process which creates polydomain regions called twins (two equivalent variant that grow one adjacent to another forming an elongated martensite needle). This kind of process can be easily understood: To balance the increment of the elastic field energy in a region near a nucleation of a martensitic domain, the closer atoms may transform to another variant of martensitic domain [4, 5]. While the domains are growing, coherent austenite/martensite interfaces appear along planes which cannot be deformed or rotated called invariant planes [6].

The final microstructure, when the phase transition is completed, is structurally inhomogeneous and may be quite complex. It consists of an apparently random combination of martensite needle shaped domains, that can be single variants of martensite or twins, which have grown from nucleation centers or from the boundaries in an avalanche-like process. The growth dynamics of this martensite-austenite interface movements has been studied with high time resolution experiments such as acoustic emission [7, 8] and high sensitivity calorimetry [9]. When cooling rate is slow enough, the system remains trapped in metastable states until free energy barrier disappears. Then, it relaxes by creating a new martensite needle, in a fast event called an avalanche. After it, the system remains again in a silent period [10]. The martensite domains are exclusive regions since the high elastic energy barriers prevent the needle plates from retransforming, thus neither coalescing nor penetrating each other [11].

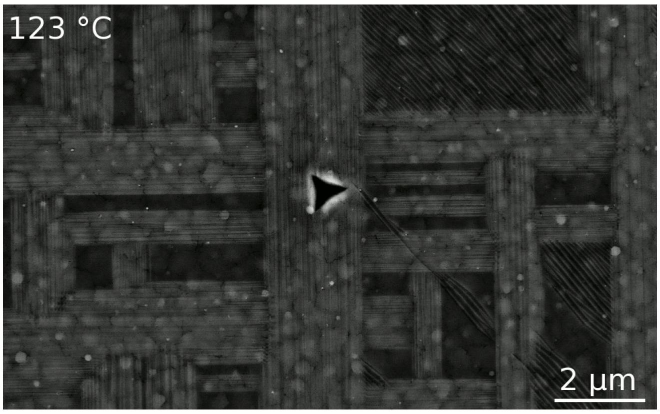

FIG. 1: Experimental image of the nucleation and growth of adaptive martensite in epitaxial layers of the shape memory alloy Ni-Mn-Ga with an indentation [12].

FIG. 1 shows an example of the experimental growth process of a microstructure of the shape memory alloy Ni-Mn-Ga, whose martensitic phase has two tetragonal variants that are favored due to the indentation in the sample, which creates the horizontal and vertical plates that can be easily distinguished from the darker austenite phase. Strictly speaking, other directions of the needles

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can also be observed (for example, at the right top of the image there are diagonal domains) because the studied sample is tridimensional, and it may have other equivalent variants.

Fourier analysis of the microstructures has revealed that the distribution of the domain sizes tends to be fat tailed (in the large size region)[7, 13]. This perfectly agrees with the statistical properties of these avalanche-like processes that tend to show absence of characteristic scales in energies, durations, sizes, etc. It has also been shown that this fat tailed distributions fits to power-laws with characteristic exponents, which shows a weak universality[10].

In this work we have studied a discrete 2D probabilistic and geometrical model proposed by Genís Torrents *et al.* [10] which assumes an elongated shape of the martensitic domains that grows in the parent matrix and the absence of retransformation events (the needles cannot cross each other)[10]. The model was simulated and analytically solved in a square system with two equivalents growth directions of the needles: horizontal (1,0) and vertical (0,1)[10]. The results revealed the existence of an effective correlation in the final microstructure between domains oriented in the same direction. We have first reproduced some of the results in reference [10] in order to check that our simulation code is correct. Secondly, we have modified the model, including a prenucleated horizontal domain in the center of the square lattice to quantitatively characterize the correlation between the transformed domains in the final microstructure.

II. MODEL AND SIMULATIONS

The proposed model is formulated on a 2D planar square lattice of dimension $L \times L$ with lattice spacing a and $L = 2^{10}$. Two equivalent growth directions are considered: horizontal (1,0) and vertical (0,1). The model considers the lattice initially in the austenite phase. The microstructure grows following these steps:

1. Using a random numbers generator (`RCARIN`) we choose a site in the lattice with uniform probability. If the site is already transformed, we choose another one that will be used for the growth of a martensitic domain.

2. With another random number we choose if the domain grows vertically or horizontally with equiprobability 1/2.

3. A linear domain is then grown in the chosen direction as far as hitting an already transformed site or the lattice boundary.

4. This three steps are sequentially repeated until the lattice is fully transformed.

At late states of the transformation, this simulation algorithm becomes more and more slow due to the low

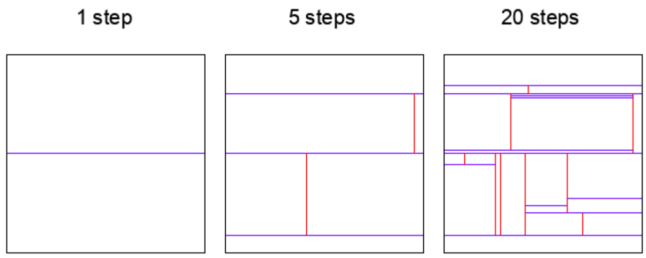

FIG. 2: Scheme of the construction of a martensitic microstructure with the proposed model which shows in purple the horizontal (0,1) direction needles and in red the vertical (1,0) ones. In each figure we can see respectively 1, 5 or 20 steps of the simulation, where we consider a step following the 3 processes that creates a domain.

probability of finding an untransformed site in the lattice just by choosing a random position. Consequently, after a certain number of steps, the algorithm is slightly modified by keeping a table of the untransformed sites and just choosing the nucleation points randomly among these. This procedure is not convenient in the beginning of the simulation because the rearrangement of the list is slow, specially for systems of large dimensions L. This code runs on a Mackbook Pro with 2.3GHz dual-core Intel Core i5 processor and needs 8.7 ± 0.8 cpu seconds for fully transform a lattice of $L = 2^{10}$. When L is changed the time scales approximately as $L^{2.62}$.

FIG. 2 shows an example of the initial steps of the construction of a microstructure following this algorithm, and FIG. 3 shows an example of the fully transformed lattice. Already in this FIG. 3 one can see that there is an effective correlation between domains that have grown in the same direction as black lines tend to be surrounded by other parallel black lines.

To prove the correctness of our code we have computed the domain size distribution for $L = 2^{10}$ and compared it with the equivalent results obtained by Genís Torrents *et al.*[10]. In the final microstructure we count how many domains ($H(\delta)$) have a linear size δ. Our data has been normalized by the number of simulation runs ($2 \cdot 10^4$) in order to get good statistics. Our result is shown in FIG. 4 together with the results from reference [10] and the exact power law. The agreement is perfect within error bars.

In order to increase our understanding of the dipole-dipole like correlations between parallel domains, the algorithm has been slightly modified: In the center of the original untransformed lattice, we have included a horizontally prenucleated domain of size d. Then, the simulation is run as explained in the original algorithm. In the final microstructure, we measure the probability $P_H(x, y)$ that a certain site at a generic position (x, y) is transformed to an horizontal domain. This probability was analyzed for different sizes of the prenucleated domain. The attention focus of the study is the behavior near the center of the lattice to avoid the finite sizes effects. In

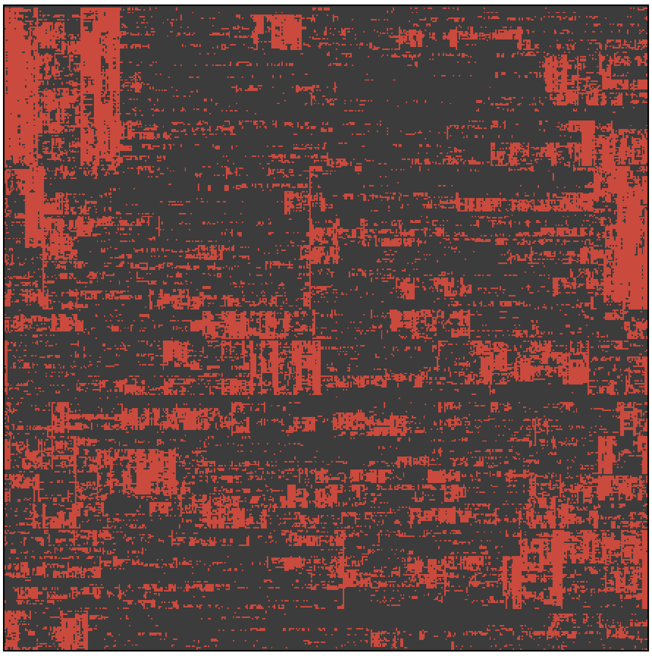

FIG. 3: Example of a final microstructure obtained with this model in which we can identify the horizontal domains painted in black and, in red, the vertical ones.

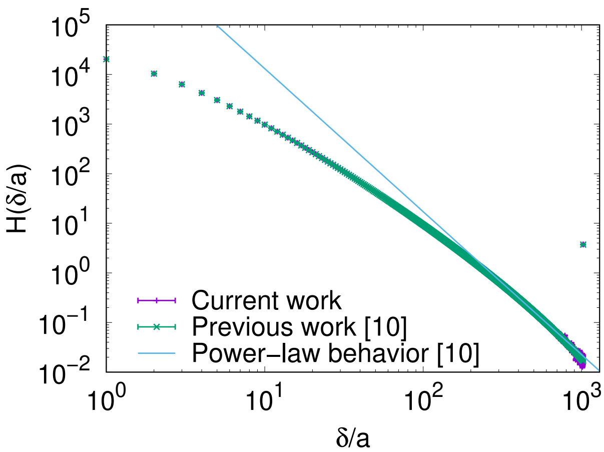

FIG. 4: Average number of domains with length δ in a system of $L = 2^{10}$. Purple dots show results from the current work, green dots show results from the previous work *et al.* [10] and the blue line the power-law behavior with exponent $\alpha = 2.882$ computed analytically in *et al.*[10] at $\delta = L/2$. Note that our results also reproduce the delta function peak at $\delta = L$ corresponding to domains of length equal to the system size.

order to compute quantitative data, we will average over the four quadrants of the lattice that are symmetrically equivalent and the origin of the (x, y) coordinates will be placed in the center of the prenucleated domain. The fact of using an initially isolated prenucleated domain in the center of the lattice has the advantage that we will separate the dipole-dipole correlations from effects due to boundary conditions. But one should notice that there is a drawback because we are introducing a domain that does not fulfill the defined growth rules since this domain has stopped its growth without touching any transformed domain, hence this may affect the points next to these limits.

III. RESULTS

Simulations have been carried for prenucleated horizontal domains of size $d = 2^6, 2^7, 2^8, 2^9$ in lattices of size $L = 2^{10}$. For each case, $1.5 \cdot 10^4$ completed microstructures have been averaged. FIG. 5 shows an example of the results corresponding to the probability $P_H(x, y)$ of having a martensitic domain in the same direction as the one which has been specifically introduced.

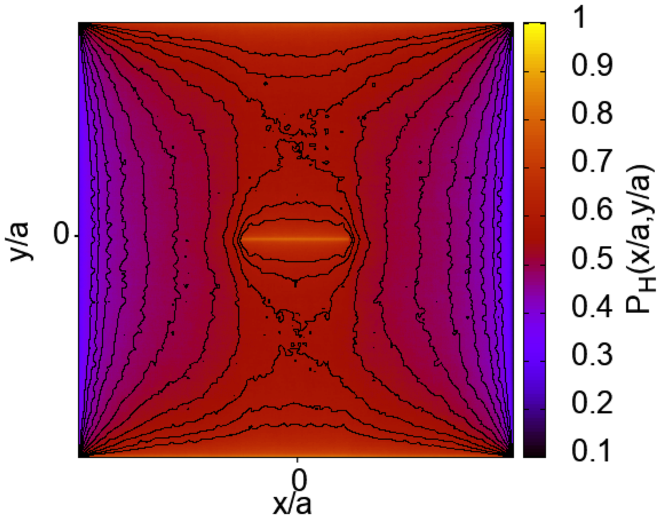

FIG. 5: Probability P_H of having an horizontal martensitic domain at site (x, y) averaged over $1.5 \cdot 10^4$ transformed microstructures of a lattice of $L = 2^{10}$. The prenucleated domain have a size $d = 2^8$. The contour lines correspond to equal probability lines. The line closer to the original domain corresponds to $P_H = 0.6$ and the rest of profiles decrease in intervals of 0.02 until $P_H = 0.4$.

As expected, in all cases it appears an excess of probability ($P_H > 0.5$) in all the neighboring sites of the prenucleated domain which attenuates as the distance increases. It also becomes pretty clear that this effective dipole-dipole interaction has a larger range in the perpendicular direction to the martensitic domain than in the parallel direction. FIG. 6 shows the behavior of the probability profile in the perpendicular direction for a fixed position in the x axis. This probability decays logarithmically as:

$$P_H(y/a) = -\beta \cdot \ln(y/a) + P_{H1}, \qquad (1)$$

This tendency diminishes drastically for perpendicular profiles which do not cross the prenucleated domain (the probability is practically constant, and its value is determined by the boundary effects of the walls). All the profiles show a peak as we get closer to the limits of the system ($y/a = 512$) due to the effects of the horizontal walls.

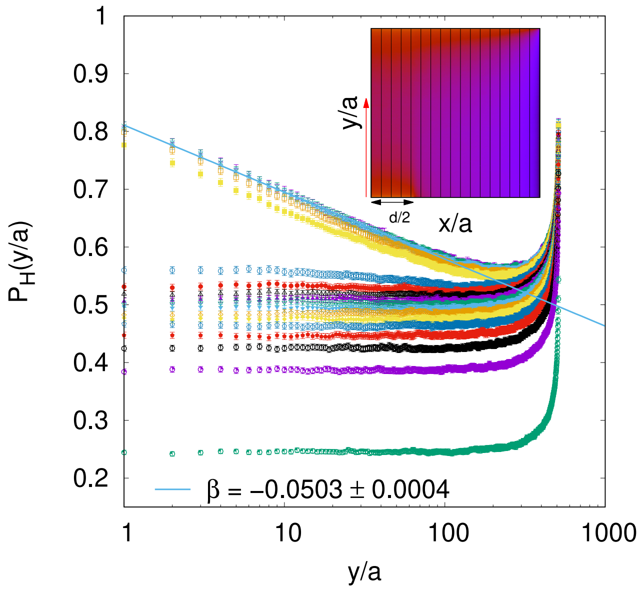

FIG. 6: Probability profiles $P_H(y/a)$ in the perpendicular direction of the prenucleated domain for different fixed x/a positions, shown every $(\Delta x)/a = 30$ sites starting at $x/a = 0$. This system has $L = 2^{10}$ and a prenucleated domain of $d = 2^8$. The inset map shows the averaged probability $P_H(x/a, y/a)$ of the four symmetrically-equivalents quadrants. Setting the origin at the center of the system, the plotted profiles start at $y/a = 1$ and finish at the boundaries (as shows the red arrow). Each profile corresponds to a vertical black line in the inset map: the top one with a logarithmic decay is fixed at $x/a = 0$, and as we move on the x axis they get lower. The double arrow d/a shows the size of the prenucleated domain. The blue line shows the logarithmic decay at $x/a = 0$ with $\beta = 0.0503 \pm 0.0004$.

FIG. 7 shows the behavior of the probability for the parallel direction to the prenucleated needle for different fixed y/a. This depicts qualitatively the drastic decay of the probability for x/a positions which are not in the range of the horizontal fixed martensitic domain.

FIG.8. And FIG. 9 show the β and P_{H1} values for every fixed x/a perpendicular probability profile $P_H(y/a)$ for four sizes of the prenucleated domain d. As it can be observed, for bigger sizes the β value increases, as well as the excess of probability in the neighboring sites P_{H1}. For all cases, the logarithmic tendency disappears for x/a bigger than the prenucleated domain sizes.

IV. CONCLUSIONS

We have studied a model for the growth of linear martensitic domains on a 2D lattice. The domains can

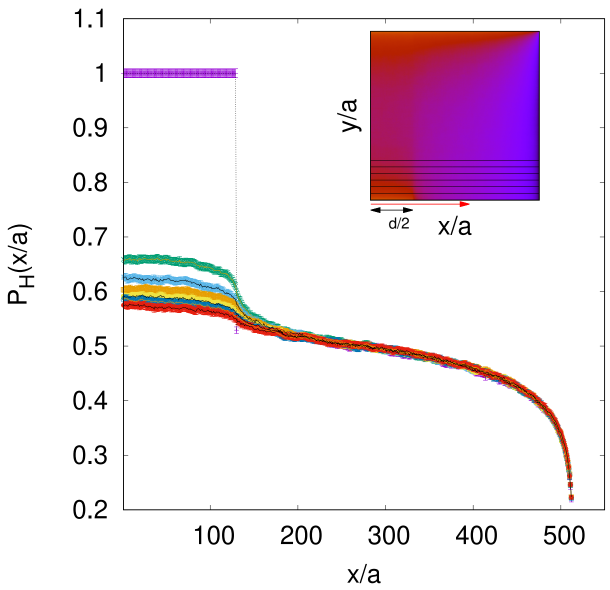

FIG. 7: Probability profiles $P_H(x/a)$ in the parallel direction of the prenucleated domain for different fixed y/a positions, shown every $(\Delta y)/a = 20$ sites starting at $y/a = 0$ and ending at $y/a = 120$. This system has $L = 2^{10}$ size and a prenucleated domain of $d = 2^8$. The inset map shows the averaged probability $P_H(x/a, y/a)$ of the four symmetrically-equivalents quadrants. Setting the origin at the center of the system, the plotted profiles start at $x/a = 1$ and finish at the boundaries (as shows the red arrow). Each profile corresponds to a horizontal black line in the inset map: the top one with a sudden decay (when the prenucleated needle ends) is the $y/a = 0$ profile, and as we move on the y axis they get lower. The double arrow d/a shows the size of the prenucleated domain. Lines are guides to the eye.

grow in two symmetrically related directions: vertical and horizontal. We have reproduced with our simulation algorithm the results in ref. [10] in order to check the correctness of our algorithm.

The novelty of this work is that we have placed a horizontal prenucleated domain in the center of the lattice in order to study the effective dipole-dipole correlations in the final microstructures. The conclusions are:

- In the neighborhood of a horizontal domain there is an excess probability ($P_H > 0.5$) of a site being occupied by a parallel (horizontal) domain.
- This probability decays logarithmically with the perpendicular distance to the prenucleated domain as $P_H(y/a) = -\beta \cdot \ln(y/a) + P_{H1}$.
- This tendency disappears drastically for the profiles with a x/a position which does not contain the prenucleated needle.
- Both β and P_{H1} values increase as the prenucleated domain gets bigger.

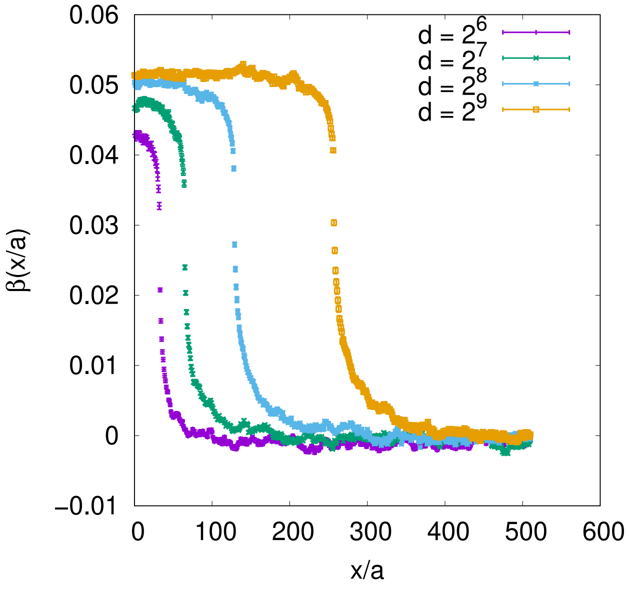

FIG. 8: Value of $\beta(x/a)$ value of the logarithmic decay for each perpendicular probability profile depending on the fixed position x/a. The plotted data corresponds to the four different d size prenucleated domains studied.

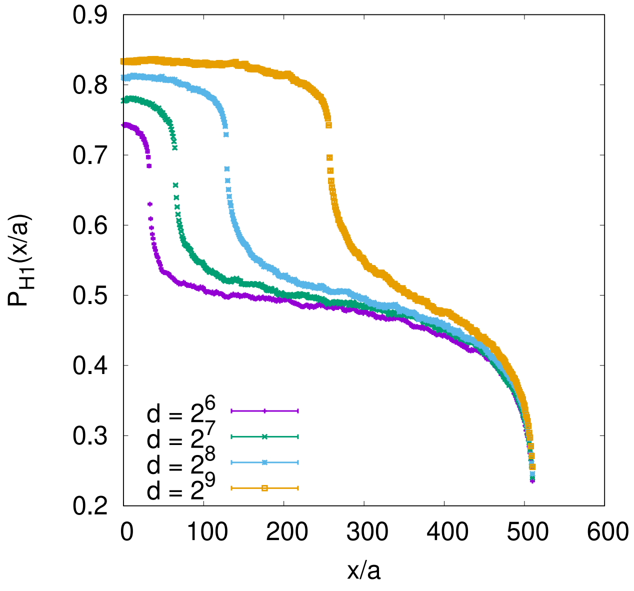

FIG. 9: Value of $P_{H1}(x/a)$ value of the logarithmic decay for each perpendicular probability profile depending on the fixed position x/a. The plotted data corresponds to the four different d size prenucleated domains studied.

To obtain the full dependence of $P_H(x, y)$ with x, y and d will require further simulations. In the future we plan to study a three-dimensional model as this may give a more realistic approach to the real behavior of this effective dipole-dipole correlation.

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[1] *Shape Memory Materials*, editet by K. Otsuka and C. M. Wayman, Cambridge University Press (Cambridge, 1998).
[2] J. W. Chritsian,*The theory of transformations in metals and alloys*, 2nd ed., Pergamon Press (Oxford, 1975).
[3] K. Bhattacharya, *Microstructure of the Martensite: why it forms and how it gives rise to the shape-memory effect*, Oxford University Press (New York, 2003).
[4] A. G. Kachaturyan, *Theory of Structural Transformations in Solids* (Dover, New York, 2008).
[5] F. J. P. Reche, *Experimentos y modelos en sistemas que presentan transiciones de fase de primer orden con dinámica de avalanchas*,Tesis Doctoral por la Universidad de Barcelona (Barcelona, 15 de marzo 2005).
[6] Z. Nishiyama, *Martensitic Transformations*, Academic Press (London, 1978).
[7] A. A. Likhachev, J. Pons, E. Cesari, A. Yu. Pasko, and V. I. Kolomytsev, Src. Mater. **43**, 765 (2000).
[8] Ll. Carrillo, Ll. Mañosa, J. Ortín, A. Planes, and E. Vives, Phys. Rev. Lett. **81**, 1889 (1998).
[9] A. Planes, Ll. Mañosa, and E. Vives, J. Alloys Comp. **577**, S699 (2013).
[10] G. Torrents, X. Illa, E. Vives, and A. Planes, Phys. Rev. E **95**, 013001 (2017).
[11] M. Rao, S. Sengupta, and H. K. Sahu, Phys. Rev. Lett. **75** (1995).
[12] R. I. Niemann, *Nukleation und Wachstum des adaptiven Martensits in epitaktischen Schichten der Formgedachtnislegierung Ni-Mn-Ga*, Doctoral dissertation (Fakultat Mathematik und Naturwissenschaften der Technischen Universitat Dresden, 2015). Retrieved from http://www.qucosa.de/fileadmin/data/qucosa/documents/18341/Dissertation_Robert_Niemann.pdf.
[13] A. Yu. Pasko, A. A. Likhachev, Yu. N. Koval, and V. I. Kolomystev, J. Phys. IV France **07** C5-435 (1997).